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Determination of actual carrier lifetime from differential measurements

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Abstract

The light-biased dynamic analysis of excess carrier decay has been known to yield differential rather than actual recombination properties since 1995. This finding implies a mismatch between actual lifetime and the measured differential lifetime in the case of injection-dependent carrier lifetime. Therefore, a time-consuming integration of differential recombination properties over the entire injection range was deemed inevitable in order to obtain actual recombination properties from differential measurements. We recently observed a striking analogy to the above finding: Time-modulated luminescence measurements – notably without any additional constant bias light – feature a systematic mismatch between actual lifetime and apparent lifetime determined from the phase shift of carrier density with respect to carrier generation if lifetime is injection-dependent. This gave rise to rearrange previous findings in a quantitative theory of light-biased carrier decay, which also comprises time-modulated lifetime techniques without constant bias light. This theory directly relates measurable differential lifetime to actual carrier lifetime. It is applied in a new *differential-to-actual* (*d2a*) lifetime analysis, which allows the determination of actual carrier lifetime from differential measurements without integration over the entire injection range. In terms of practical relevance, the *d2a* approach brings about a drastic experimental simplification. Combined with the general advantages of dynamic lifetime techniques, it could upgrade differential carrier decay techniques and time-modulated lock-in techniques into powerful quantitative characterization options in silicon photovoltaics. This paper provides an experimental proof of concept of the *d2a* technique based on harmonically time-modulated photoluminescence.

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1. Introduction

Carrier lifetime measurements based on light-biased excess carrier decay analyses had extensively been used in the past. Back in the mid-nineties however, Brendel, Aberle, and Schmidt identified a discrepancy between the decay times obtained via such analysis and actual carrier lifetime [1-3]. Light-biased carrier decay analyses were found to yield differential rather than actual recombination properties – leading to a systematic mismatch in the case of injection-dependent carrier lifetime. Therefore, integration of such differential recombination properties over the entire injection range below a given injection level of interest was deemed inevitable in order to determine actual recombination properties [2-4].

In the course of our investigation of lifetime measurements via harmonically time-modulated luminescence, we observed a striking analogy to the findings of Brendel, Aberle, and Schmidt. The phase shift between excess carrier generation rate G and excess carrier density Δn proved to be greater than effective carrier lifetime τ in the case of a positive derivative $d\tau/d\Delta n > 0$, and vice versa. This observation gave rise to rearrange the previous findings in a quantitative theory of light-biased carrier decay [5], which also encompasses time-modulated lifetime techniques without any constant bias light [6-10]. This theory quantitatively relates differential lifetime to actual carrier lifetime τ through its derivative $d\tau/d\Delta n$.

Based on the quantitative theoretical description of light-biased dynamic measurements of effective carrier lifetime given in [5], this paper provides an experimental proof of concept of the derivative lifetime technique *d2a (differential-to-actual)* via harmonically time-modulated photoluminescence. Further, this paper elaborates whether or not previously addressed constraints associated with a non-uniform depth distribution of excess carrier density and generation rate [2,3] affect the *d2a* lifetime technique.

2. Theory of light-biased decay time

In this paper, we give a brief and concise outline of the theory of light-biased decay time. An elaborate account of this theory is to be found in [5].

The dynamics of excess carriers in silicon is usually adequately described by the time-dependent continuity equation

$$\frac{d\Delta n(t)}{dt} + \frac{\Delta n(t)}{\tau} = G(t), \quad (1)$$

with excess carrier density Δn , actual effective carrier lifetime τ , and excess carrier generation rate G . Assuming an idealized abrupt change of excess carrier generation rate at $t = 0$ such that $G(t \leq 0) = G_0$ and $G(t > 0) = \xi G_0$ with a bias light parameter $0 \leq \xi < 1$, for a positive time $t \rightarrow 0$ one obtains

$$\frac{d\Delta n(t \rightarrow 0)}{dt} = -G_0 \cdot (1 - \xi). \quad (2)$$

With an initial excess carrier density $\Delta n_0 = \tau_0 G_0$, the measurable decay time τ_m is

$$\tau_m(t \rightarrow 0) = \frac{\Delta n_0 - \Delta n(t \rightarrow \infty)}{(1 - \xi)G_0}. \quad (3)$$

This equation reveals the differential nature of measurable decay time as the ratio between a change of

excess carrier density and a change of generation rate. With a first order series expansion of actual carrier lifetime $\tau(\Delta n) \approx \tau_0 + (\Delta n - \Delta n_0) \cdot d\tau/d\Delta n$, the term $\Delta n(t \rightarrow \infty)$ reads

$$\Delta n(t \rightarrow \infty) = \xi \Delta n_0 \frac{1 - G_0 \left. \frac{d\tau}{d\Delta n} \right|_{\Delta n_0}}{1 - \xi G_0 \left. \frac{d\tau}{d\Delta n} \right|_{\Delta n_0}}. \quad (4)$$

This allows expression of the actual carrier lifetime as a function of the measurable decay time (differential lifetime):

$$\tau_0 = \tau_m \cdot \left(1 - \xi G_0 \left. \frac{d\tau}{d\Delta n} \right|_{\Delta n_0} \right) \quad (5)$$

In the limit $\xi \rightarrow 1$ representative of continuously (e.g. harmonically) time-modulated lifetime techniques, it can be shown that this relation is valid and exact [5], regardless of the curvature of $\tau(\Delta n)$. This relation is very accurate for values of $\xi > 0.9$ which are typical for conventional (small signal) light-biased excess carrier decay time measurements [1-4].

3. Implications

Eq. (3) reveals the differential nature of decay time as the ratio between a change of excess carrier density and a change of excess carrier generation rate. For changes of generation rate with $\xi > 0$ and at a finite derivative of actual lifetime with respect to excess carrier density, the relative changes of generation rate and excess carrier density cannot coincide. Therefore, the measured decay time must differ from actual carrier lifetime.

3.1. Limits

From the essential Eq. (5), it can be concluded that in the trivial cases $d\tau/d\Delta n = 0$ and $\xi = 0$, differential lifetime τ_m and actual lifetime τ_0 coincide. Therefore, excess carrier decay time measurements without bias light yield the actual lifetime.

With finite bias light parameters ξ and at a non-negligible injection dependence of carrier lifetime, the discrepancy between differential lifetime and actual lifetime increases with increasing ξ , to where it becomes most pronounced in the limit $\xi \rightarrow 1$. Notably, this dependence on the bias light parameter ξ has been neglected in previous works on the topic [1-4], assuming a maximal discrepancy between differential and actual lifetime (corresponding to $\xi \rightarrow 1$) even though *small signal* decay time techniques typically feature bias light parameters in the range of 0.9-0.95.

3.2. Continuously time-modulated lifetime techniques

A continuous (e.g. harmonic) time modulation of excess carrier generation rate can be perceived as a time sequence of infinitesimal changes of generation rate. It is therefore representative of the limit $\xi \rightarrow 1$ in the above treatment. In quasi-steady-state, the superposition of excess carrier decays caused by such changes of generation rate is a total excess carrier density whose maximum is shifted by differential

lifetime τ_m with respect to the maximum of excess carrier generation rate. A rigorous proof of this is also stated in [5].

With the above perception, the time shift between excess carrier density and harmonically time-modulated generation rate corresponds to differential rather than actual carrier lifetime, regardless of an additional steady-state light bias.

It should be noted that Eq. (5) also accords with previous quantitative statements about the discrepancy between differential lifetime and actual lifetime, such as Schmidt's prediction of $3\tau_m = \tau_0$ for the Auger limit [3], which was at that time excellently confirmed by the mismatch between MFCA [6] and QSSPC [11] measurements.

4. The $d2a$ lifetime technique

4.1. Working principle

Eq. (5) gives access to actual lifetime from a combination of at least two measurements of differential lifetime, to be conducted at adjacent generation rates. We refer to this lifetime technique as $d2a$ (*differential-to-actual*). If linearity of lifetime as a function of Δn (constant $d\tau/d\Delta n$) is locally satisfied at an injection level of interest, two adjacent measurements suffice. Two measured excess carrier generation rates G_i and corresponding differential lifetimes $\tau_m(G_i)$ can be inserted in the system of equations ($i = 1, 2$)

$$\tau(G_i) = \tau_m(G_i) \cdot \left(1 - \xi G_i \left. \frac{d\tau}{d\Delta n} \right|_{\Delta n_i} \right). \quad (6)$$

Assuming a constant $d\tau/d\Delta n$, the actual lifetimes can be expressed in terms of each other and the system of equations can be solved.

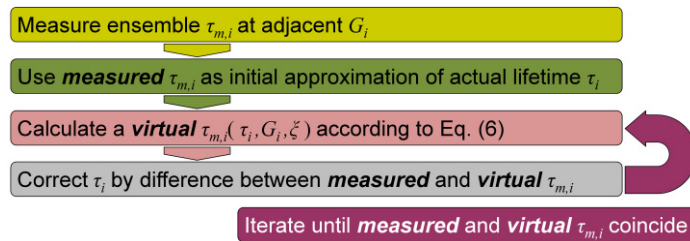


Fig. 1 Flow chart of the working principle of the $d2a$ lifetime analysis. Here, the index i refers to a differential lifetime measurement at the excess carrier generation rate G_i

For nonlinear lifetime, Eq. (6) allows an iterative determination of actual lifetime from a set of more than two measurements of differential lifetimes at adjacent generation rates (cf. flow chart in Fig. 1). We had previously proposed a related carrier lifetime approach based on harmonically time-modulated photoluminescence [9,10]. The iteration method proposed therein can be adapted here. Yet, at that time we related actual lifetime to differential lifetime through the numerical (finite-element) solution of the continuity equation. This step can now be replaced by the analytic relation given in Eqs. (5) and (6), respectively.

It can be shown that a non-uniform depth distribution of excess carrier density does not affect the $d2a$ analysis (cf. Appendix A).

4.2. Validation via simulated excess carrier density

One option to validate the $d2a$ technique is the lifetime analysis of accurate numerical (finite-element) simulations of excess carrier density $\Delta n(t)$ e.g. in a harmonically time-modulated experiment. We applied the $d2a$ analysis to a highly nonlinear lifetime curve. Based on a true lifetime curve $\tau(\Delta n)|_{\text{true}}$ resembling typically encountered curve shapes (cf. Fig. 2), we first modeled the excess carrier density $\Delta n(t)$ to be expected upon a harmonically time-modulated quasi-steady-state excess carrier generation rate $G(t)$. Subsequently, we determined differential lifetimes $\tau_m(G)$ as injection-dependent time shifts between generation rate and excess carrier density, as proposed in [10] (depicted in Fig. 2 as yellow triangles). These differential lifetimes were finally analyzed via the $d2a$ technique and compared to true lifetime, yielding a convincing agreement as can be seen from Fig. 2.

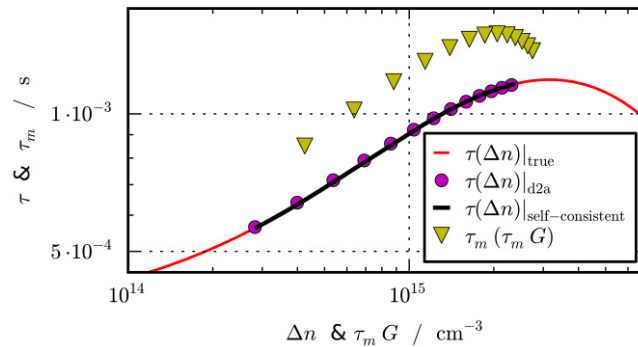


Fig. 2 $d2a$ lifetime analysis of simulated differential lifetimes τ_m as expected from a true lifetime curve $\tau(\Delta n)|_{\text{true}}$ upon harmonic time modulation of excess carrier generation $G(t)$. A self-consistent lifetime analysis [12] of the simulation result is also shown.

4.3. Experimental validation

A complementary option is to validate the $d2a$ technique experimentally. For this purpose, we applied the $d2a$ technique to injection-dependent time-shifts obtained from a harmonically time-modulated quasi-steady-state photoluminescence measurement on a 160 μm thick 1 Ωcm silicon wafer. The resulting injection-dependent actual effective carrier lifetime is plotted in Fig. 3.

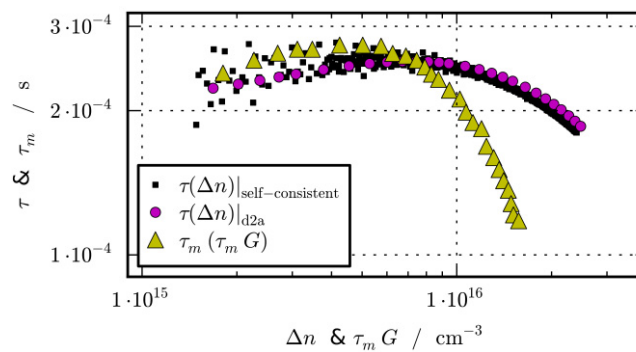


Fig. 3 Experimental validation of the $d2a$ lifetime technique via time-modulated quasi-steady-state photoluminescence. Differential lifetime τ_m considerably differs from actual lifetime (here represented by a self-consistent lifetime analysis [12]), but the $d2a$ analysis coincides with the self-consistent lifetime analysis.

As we do not a priori know the true lifetime in this experimental case, the resulting lifetime is compared to a self-consistent lifetime analysis [12] which is also shown in Fig. 3. As the self-consistent lifetime analysis applied to the noise-free simulated data in Fig. 2 coincides with true lifetime, this is to be expected from the experimental case as well. As can be seen in Fig. 3, self-consistent and $d2a$ results virtually coincide. The residual minor mismatch could be caused by statistical noise or by a small systematic error of the assumed dopant concentration, which is required for the self-consistent lifetime analysis.

5. Conclusion

Based on a recently published theory of light-biased decay time [5], this paper provides an experimental proof of concept of the $d2a$ lifetime technique derived from this theory. Further, it is reasoned why previously addressed constraints associated with a non-uniform depth distribution of excess carrier density and generation rate [2,3] are practically irrelevant to the $d2a$ lifetime technique.

For conventional light-biased excess carrier decay analyses, the $d2a$ technique enables the determination of actual carrier lifetime from differential lifetime measurements without integration over the entire injection range – thereby substantially reducing measurement uncertainty and experimental effort. For continuously time-modulated and purely phase-sensitive (possibly lock-in-based) lifetime techniques [6-9], the $d2a$ technique gives access to actual rather than differential lifetime in the first place. Further, it opens up a pathway toward highly sensitive and accurate lock-in-based measurement techniques of actual lifetime beyond the state-of-the-art self-consistent carrier lifetime analysis [12], which relies on the time-dependent recording of excess carrier density rather than on straightforward phase information.

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Appendix A. Non-uniform depth distribution of excess carrier density and generation rate

Both Schmidt [3] as well as Schuurmans *et al.* [4] have previously addressed errors resulting from the integration of differential lifetimes in the case of a non-uniform depth distribution of excess carrier density and generation rate. Non-uniform depth distributions occur if wafer thickness d significantly exceeds both light penetration depth and minority carrier diffusion length. As lifetime analyses based on Eq. (1) are usually based on depth-averaged excess carrier density and generation rate, both quantities are effectively underestimated in the case of interest due to a reduced electronically active effective substrate thickness $d^* < d$. This could be taken into account in a depth-averaged treatment by introducing a factor $\kappa = d^*/d > 1$. The continuity equation would then read

$$\frac{d\Delta n(t)}{dt} + \frac{\Delta n(t)}{\tau} = G(t) \cdot \kappa. \quad (\text{A.1})$$

With this enhanced depth-averaged excess carrier generation rate $G \rightarrow G \cdot \kappa$, the resulting excess carrier density according to Eq. A.1 is also enhanced by the same factor $\Delta n \rightarrow \Delta n \cdot \kappa$. Therefore, differential lifetime τ_m as of Eq. (3) is independent of κ . The same applies to actual lifetime τ_0 as of Eq. (5) due to the fact that the derivative $d\tau/d\Delta n$ is accordingly reduced to $d\tau/d\Delta n \rightarrow 1/\kappa \cdot d\tau/d\Delta n$.

The $d2a$ analysis is thus unaffected by a possibly non-uniform depth distribution of excess carrier density and generation rate. Yet, the determination of the depth-averaged excess carrier density of interest would be erroneous, albeit corrigible through an intensity-weighted averaging of excess carrier density as e.g. proposed in [13].